Non-ergodic transitions in many-body Langevin systems: a method of dynamical system reduction

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Abstract. We study a non-ergodic transition in a many-body Langevin system. We first derive an equation for the two-point time correlation function of density fluctuations, ignoring the contributions of the third- and fourth-order cumulants. For this equation, with the average density fixed, we find that there is a critical temperature at which the qualitative nature of the trajectories around the trivial solution changes. Using a method of dynamical system reduction around the critical temperature, we simplify the equation for the time correlation function into a two-dimensional ordinary differential equation. Analyzing this differential equation, we demonstrate that a non-ergodic transition occurs at some temperature slightly higher than the critical temperature.

1. Introduction

Glassy systems exhibit distinctive phenomena, such as divergence of the viscosity [1], a history-dependent response [2], and an extreme slowing down of the time correlation of density fluctuations [3, 4]. These phenomena have been studied extensively through laboratory experiments, numerical experiments, and theoretical analyses of models [5]. Although each of these phenomena has gradually come to be understood individually, we are still lacking a unified picture that can account for all of them. In contrast to the rich variety of physical phenomena displayed by these systems, the types of theoretical methods that have been used in their investigation are few. Particularly, many analyses are based on the mode coupling theory (MCT) [3]. However, the MCT is not justified completely from a microscopic viewpoint, and also it is not regarded as the first-order description of a systematic formulation. Therefore, it is important to formulate a new method for analyzing the phenomena of glassy systems.

Recalling the historical development of the theory of glassy systems, we realize that the MCT has been the most commonly used theory [6] because it describes various phenomena on the basis of the singular behavior of the two-point time correlation function of density fluctuations. This time correlation function exhibits a two-step relaxation as a function of the time difference and a plateau regime appears between the two steps in the relaxation. The MCT predicts the transition temperature for a fixed density (or the transition density for a fixed temperature) at which the plateau regime extends to infinity. This phenomenon is called a *non-ergodic transition*. Although the singularity might be an artifact of the approximation employed in the theory [6], there are evidences suggesting that the non-ergodic transition has been observed experimentally and numerically with fairly good quantitative correspondence to the theoretical prediction [4]. Noting such evidence, in the present paper, we theoretically study the behavior of the time correlation of density fluctuations.

Let us briefly review the essence of the MCT. This theory provides a self-consistent integral equation of the time correlation function and the response function, ignoring vertex corrections [3]. Then, by analyzing this equation numerically, one can find a non-ergodic transition. Here, it should be noted that the memory contribution to the equation for the time correlation function is expressed in terms of the time correlation function. It is believed that such non-linear memory plays a key role in the non-ergodic transition.

Now, it is interesting that behavior similar to that exhibited in the non-ergodic transition seen in glassy systems is easily obtained in ordinary differential equations possessing no nonlinear memory. As an example, let us consider Newton's equation of motion for a point particle moving in a one-dimensional space. Let $x(t)$ be its position and $V(x)$ be a potential function given by $V(x) = -x^2\left[(x - x_*)^2 + \varepsilon\right]$, with $\varepsilon \geq 0$. We consider the particle motion under the condition that $x \to 0$ as $t \to \infty$. With this condition, it is easily found that when ε is small, the particle starting from $x(0)$ $(> x_*)$ climbs the potential, slowly passes the non-zero maximum position, and then converges

to $x = 0$. The graph of x as a function of t exhibits a two-step relaxation behavior with a long plateau. Furthermore, if $\varepsilon = 0$, the particle converges to the position x_* and cannot approach the origin. This qualitative change in the behavior of the system as a function of ε occurring at $\varepsilon = 0$ is analogous to that exhibited by glassy systems at the non-ergodic transition.

Clearly, this simple Newtonian system has no direct relation with glassy systems. Nevertheless, it might be possible that there exists a variable representing the time correlation function for a glassy system that obeys an equation similar to that in the example discussed above. Such a variable would provide a characterization of the non-ergodic transition in that glassy system. Beginning from this speculation, in this paper, we determine such a variable and derive its evolution equation from a many-body Langevin model, employing several assumptions. We demonstrate that the obtained equation indeed does exhibit a non-ergodic transition.

2. Model

We investigate a system consisting of N identical Brownian particles suspended in a liquid. We denote the volume of the system by V and the temperature by T, and consider the thermodynamic limit, represented by $V \to \infty$ and $N \to \infty$, with the density $\bar{\rho} = N/V$ fixed. Let $\boldsymbol{r}_i(t)$ be the position of the i-th particle, with $i = 1, 2, \cdots N$. We assume that the i-th particle interacts with the j-th particle through some interaction potential $U(|\boldsymbol{r}_i - \boldsymbol{r}_j|)$. The motion of each particle is described by a Langevin equation with a friction constant γ. Then, the evolution equation of the fine-grained density field, $\rho(\boldsymbol{r}, t) \equiv \sum_{i=1}^{N} \delta(\boldsymbol{r} - \boldsymbol{r}_i(t))$, is given by

$$\frac{\partial \rho(\boldsymbol{r}, t)}{\partial t} = \boldsymbol{\nabla} \cdot \left[\frac{1}{\gamma} \int d^3\boldsymbol{r}' \rho(\boldsymbol{r}', t) \rho(\boldsymbol{r}, t) \boldsymbol{\nabla} U(|\boldsymbol{r} - \boldsymbol{r}'|) + \frac{T}{\gamma} \boldsymbol{\nabla} \rho(\boldsymbol{r}, t) + \sqrt{\frac{2T}{\gamma} \rho(\boldsymbol{r}, t)} \boldsymbol{\Xi}(\boldsymbol{r}, t) \right], \tag{1}$$

where $\boldsymbol{\Xi} (= (\Xi_x, \Xi_y, \Xi_z))$ is zero-mean Gaussian white noise that satisfies

$$\langle \Xi_\alpha(\boldsymbol{r}, t) \Xi_\beta(\boldsymbol{r}', s) \rangle = \delta_{\alpha\beta} \delta(t - s) \delta(\boldsymbol{r} - \boldsymbol{r}'). \tag{2}$$

This evolution equation can be derived exactly from the Langevin equation describing the motion of N particles [7]. Explicitly, we choose the form of the interaction potential as

$$\hat{U}(k) = \frac{E}{\lambda_1 (k^2 + \lambda_1^2)} - \frac{2kE\sigma}{(k^2 + \lambda_2^2)^2}, \tag{3}$$

where $k = |\boldsymbol{k}|$. Throughout this paper, $\hat{f}(\boldsymbol{k})$ is used to represent the Fourier transform of $f(\boldsymbol{r})$: $\hat{f}(\boldsymbol{k}) = \int d^3\boldsymbol{r} \mathrm{e}^{i\boldsymbol{k}\cdot\boldsymbol{r}} f(\boldsymbol{r})$. All quantities in this system are converted into dimensionless forms by setting $\lambda_1 = E = \gamma = 1$. Further, for the specific example that we analyze numerically, we consider the case in which $\sigma = 8.0$ and $\lambda_2 = \lambda_1$. With

this choice, $U(r)$ has a minimum value -1.18 at $r = 0.09$, while $U(r) \to \infty$ as $r \to 0$ and $U(r) \to 0$ as $r \to \infty$. Explicitly one can obtain

$$U(r) = \frac{\mathrm{e}^{-r}}{4\pi r} - \frac{4}{\pi^2}\left(1 + \frac{r}{2}\frac{d}{dr}\right)\frac{\mathrm{e}^{-r}\mathrm{Ei}(r) - \mathrm{e}^{r}\mathrm{Ei}(-r)}{r}, \tag{4}$$

where $\mathrm{Ei}(r)$ represents an integral exponential function defined as $\mathrm{Ei}(r) = \int_{-\infty}^{r} dt \mathrm{e}^t/t$. We also fix the density $\bar{\rho}$ as $\bar{\rho} = 4.436$. The temperature T is regarded as a control parameter.

For later convenience, we define the quantity $\Psi(\boldsymbol{k}, \boldsymbol{k}'; t, s) \equiv \hat{\delta}\rho(\boldsymbol{k}, t)\hat{\delta}\rho(\boldsymbol{k}', s)/\bar{\rho}^2$, where $\delta\rho(\boldsymbol{r}, t) \equiv \rho(\boldsymbol{r}, t) - \bar{\rho}$. The statistical average of $\Psi(\boldsymbol{k}, \boldsymbol{k}'; t, s)$ provides the two-point correlation function $\hat{C}(k, t)$ according to the relation

$$\langle \Psi(\boldsymbol{k}, \boldsymbol{k}'; t, s) \rangle = (2\pi)^3 \hat{C}(k, t - s)\delta(\boldsymbol{k} + \boldsymbol{k}'), \tag{5}$$

where we have assumed that the statistical properties of density fluctuations are symmetric with respect to spatial translations, rotations, and temporal translations. Although the last assumption is not valid below the glass transition temperature, we focus on the higher temperature regime, in which stationarity holds. Now, following the motivation of this study, we wish to obtain a differential equation that determines the time dependence of the correlation function $\hat{C}(k, t)$. For this purpose, as a trial, let us first consider the quantity $\partial^2\hat{C}(k, t)/\partial t^2$. This quantity can be expressed in terms of $\hat{C}(k, t)$ and a four-point correlation function. Then, writing this equality, we obtain an evolution equation for $\hat{C}(k, t)$ by making the simplest approximation of omitting the third- and fourth-order cumulant terms of this four-point correlation function. The equation obtained with this approximation represents a mean field theory in the sense that fluctuation effects of Ψ are ignored when we consider its average value. As far as we know, this naive approximation was not employed in previous studies. It might be important to find some connection with more traditional approximations such as a Kirkwood superposition approximation. With this approximation, we obtain the following evolution equation for $\hat{C}(k, t)$:

$$\begin{aligned}
\frac{\partial^2\hat{C}(k, t)}{\partial t^2} &= \left(\frac{\bar{\rho}}{\gamma}\hat{U}(k) + \frac{T}{\gamma}\right)^2 k^4\hat{C}(k, t) \\
&+ \frac{\bar{\rho}^2}{\gamma^2}\int \frac{d^3\boldsymbol{k}_1}{(2\pi)^3}\hat{C}(k_1, t)\hat{C}(|\boldsymbol{k} - \boldsymbol{k}_1|, t) \\
&\left[\hat{U}(k_1)^2\left(\boldsymbol{k}\cdot\boldsymbol{k}_1\right)^2 + \hat{U}(k_1)\hat{U}(|\boldsymbol{k} - \boldsymbol{k}_1|)\left(\boldsymbol{k}\cdot\boldsymbol{k}_1\right)\left(\boldsymbol{k}\cdot(\boldsymbol{k} - \boldsymbol{k}_1)\right)\right],
\end{aligned} \tag{6}$$

with $t > 0$. Here, we impose the boundary conditions $\lim_{t\to\infty}\hat{C}(k, t) = 0$ and $\partial_t\hat{C}(k, t)|_{t=0+} = -k^2T\bar{\rho}/\gamma$, which can also be derived from (1).

Here, it is expected that $\hat{C}(k, t)$ decays rapidly to zero in the limit $k \to \infty$ and exhibits no singularity as a function of k for any t. Therefore, we can expand $\hat{C}(k, t)$ in terms of the set of Hermite functions $\phi_n(k)$, with $n = 0, 1, \cdots$ as $\hat{C}(k, t) = \sum_{n=0}^{\infty} C_n(t)\phi_n(k)$. Then, (6) can be rewritten as

$$\sum_{m=0}^{\infty} G_{nm}\frac{dC_m(t)}{dt} = B_n(t), \tag{7}$$

$$\sum_{m=0}^{\infty} G_{nm} \frac{dB_m(t)}{dt} = \sum_{m=0}^{\infty} C_m(t) L_{nm} + \sum_{m,l=0}^{\infty} M_{nml} C_m(t) C_l(t). \tag{8}$$

Here, we have introduced the new variable $B_n(t)$ in order to obtain a first-order differential equation. The coefficients L_{nm} and M_{nml} in (8) are determined by the system parameters. For example, L_{nm} is calculated as

$$L_{nm} = \frac{\bar{\rho}^2}{\gamma^2} \int_{-\infty}^{\infty} dk \phi_n(k) \phi_m(k) \left(\hat{U}(k) + \frac{T}{\bar{\rho}} \right)^2, \tag{9}$$

while G_{nm} in (7) and (8) is given by

$$G_{nm} = -\int_{-\infty}^{\infty} dk \frac{1}{k^2 + \Lambda^2} \phi_n(k) \phi_m(k), \tag{10}$$

where we have introduced a cutoff parameter Λ in order to allow expansion of the inverse of the Laplacian in terms of the Hermite functions. We take the limit $\Lambda \to 0$ after the transition temperature is calculated for a system with Λ fixed. In the actual computations, checking the temperature in the cases $\Lambda^2 = 10^{-2}$, 10^{-3}, and 10^{-4}, we found that $\Lambda^2 = 10^{-3}$ is a sufficiently small value. The numerical values appearing in the analysis hereafter were obtained using $\Lambda^2 = 10^{-3}$.

Now we describe a further approximation. Setting

$$\boldsymbol{u} = (C_0, C_1, \cdots, C_{K-1}, B_0, \cdots, B_{K-1}), \tag{11}$$

we approximate (7) and (8) by the following ordinary differential equation:

$$\hat{G} \frac{d\boldsymbol{u}}{dt} = \hat{\Sigma} \boldsymbol{u} + \hat{\Theta}(\boldsymbol{u}, \boldsymbol{u}). \tag{12}$$

Here, the $2K \times 2K$ matrices $\hat{G}$ and $\hat{\Sigma}$ are defined as $\hat{G}_{nm} = \hat{G}_{K+n,K+m} = G_{nm}$ and $\hat{\Sigma}_{n,K+m} = \delta_{nm}$, $\hat{\Sigma}_{K+n,m} = L_{nm}$, for $1 \le n, m \le K$ (The other components are zero). Also, $\hat{\Theta}$ is determined from M_{nml} and represents a map from two copies of $\boldsymbol{u}$ to a vector. We chose $K = 100$ for the analysis reported below. (We checked the K dependence of L_{nm}, G_{nm} and M_{nml}, and confirmed that $K = 100$ is sufficiently large.)

3. Analysis

We investigate the system behavior as we change the temperature T. We first focus on the behavior of solution trajectories of (12) near the origin, $\boldsymbol{u} = 0$. When $|\boldsymbol{u}|$ is sufficiently small, this behavior is approximated by the linear equation obtained by omitting the nonlinear term in (12). Then, calculating the eigenvalues and eigenvectors of $\hat{\Sigma}$, we can classify the solution trajectories. In the high temperature limit, from the form of L_{nm}, it is easily found that all trajectories either approach or move away from the origin exponentially, because the eigenvalues consist of K pairs of positive and negative numbers. We found in the numerical computations that when the temperature is decreasing there exists a temperature T_0 below which one pair of eigenvalues becomes zero. (We have T_0=19.535 in the example we study.) Thus, the solution trajectories in the $2K$-dimensional phase space undergo qualitative change when the temperature

passes through T_0. The corresponding eigenvectors at T_0, which we denote $\mathbf{\Phi}_{00}$ and $\mathbf{\Phi}_{01}$, play a prominent role in the subsequent analysis.

It is known that this type of qualitative change near $T = T_0$ can be described by a nonlinear differential equation for the amplitudes of these zero eigenvectors [8]. We now attempt to derive such an equation. Let $\hat{\Sigma}_0$ be $\hat{\Sigma}$ at $T = T_0$. Then, we find that $\mathbf{\Phi}_{00}$ and $\mathbf{\Phi}_{01}$ satisfy the relations

$$\hat{\Sigma}_0 \mathbf{\Phi}_{00} = 0, \tag{13}$$

$$\hat{\Sigma}_0 \mathbf{\Phi}_{01} = \mathbf{\Phi}_{00}. \tag{14}$$

Now, setting $\epsilon = (T - T_0)/T_0$, we consider the solution trajectories satisfying $\mathbf{u} \to \mathbf{0}$ in the limit $t \to \infty$ for the system with small positive ϵ. For such trajectories, we expand $\boldsymbol{u}$ in eigenvectors of $\hat{\Sigma}_0$ as

$$\boldsymbol{u}(t) = A_1(t)\epsilon \mathbf{\Phi}_{00} + A_2(t)\epsilon^{3/2}\mathbf{\Phi}_{01} + \boldsymbol{\eta}(A_1(t), A_2(t)), \tag{15}$$

where $\boldsymbol{\eta}$ represents the contribution to $\boldsymbol{u}$ that is not from the zero eigenvectors, and the amplitudes of the other eigenvectors quickly decay to the values determined by A_1 and A_2. Then, the time evolution of the variables A_1 and A_2 is described by autonomous equations of the form

$$\frac{dA_i(t)}{dt} = \Omega_i(A_1(t), A_2(t)). \tag{16}$$

Now, substituting (15) into (12) and using expansions

$$\boldsymbol{\eta} = \epsilon^{3/2}\boldsymbol{\eta}^{(3/2)} + \epsilon^2\boldsymbol{\eta}^{(2)} + \epsilon^{5/2}\boldsymbol{\eta}^{(5/2)} + \cdots, \tag{17}$$

$$\Omega_i = \epsilon^{1/2}\Omega_i^{(1/2)} + \epsilon^1\Omega_i^{(1)} + \epsilon^{3/2}\Omega_i^{(3/2)} + \cdots, \tag{18}$$

we can determine the functional forms of $\boldsymbol{\eta}(A_1, A_2)$ and $\Omega_i(A_1, A_2)$ using a perturbative method. Note that the dependences on ϵ in (15), (17), and (18) are chosen so that the perturbative expansion can be carried out in a systematic manner. For example, all the terms proportional to $\epsilon^{3/2}$ in the expression obtained with the above procedure yield

$$\hat{G}\mathbf{\Phi}_{00}\Omega_1^{(1/2)}(A_1, A_2) = A_2\mathbf{\Phi}_{00} + \hat{\Sigma}_0\boldsymbol{\eta}^{(3/2)}(A_1, A_2). \tag{19}$$

Because $\hat{\Sigma}_0$ has a zero eigenvalue, it is not invertible. Thus, when we regard (19) as a linear equation for $\boldsymbol{\eta}^{(3/2)}$, there are either no solutions or an infinite number of solutions. In order to carry out the perturbative calculation systematically, we impose the solvability condition under which $\boldsymbol{\eta}^{(3/2)}$ can be obtained uniquely. This solvability condition determines $\Omega_1^{(1/2)}$. Applying similar procedures to each of the equations appearing successively, which are proportional to ϵ^2, $\epsilon^{5/2}$, $\cdots$, we derive $\Omega_1^{(1)}$, $\Omega_2^{(1/2)}$, $\Omega_1^{(3/2)}$, $\Omega_2^{(1)}$, $\cdots$ in an iterative manner. Combining the contributions up to $\Omega_1^{(2)}$ and $\Omega_2^{(3/2)}$, we obtain

$$\frac{dA_1}{dt} = aA_2, \tag{20}$$

$$\frac{dA_2}{dt} = b_1A_1 + b_2A_1^2 + b_3A_1^3 + b_4A_2^2, \tag{21}$$

where each coefficient can be calculated as a function of T. For the example we analyzed numerically, we found $a = -0.061186\epsilon^{1/2}$, $b_1 = -1.108178\epsilon^{3/2}$, $b_2 = -0.000566\epsilon^{1/2}$, $b_3 = -0.011904\epsilon^{3/2}$ and $b_4 = 0.836102\epsilon^{3/2}$. (The details of the calculation will be reported in another paper [9].)

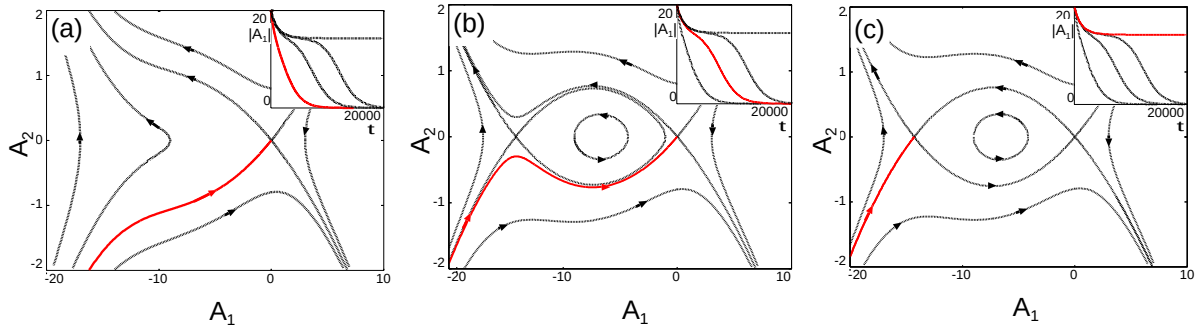

Figure 1. Solution trajectories of (20) and (21) in (A_1, A_2) space for three values of ϵ. (a) $\epsilon = 2.520 \times 10^{-3}$, (b) $\epsilon = 2.294 \times 10^{-3}$, and (c) $\epsilon = 2.285 \times 10^{-3}$. The red curves represent physical solutions that satisfy $\lim_{t\to\infty} A_i = 0$. Inset: A_1 as a function of t. The red curves in the insets correspond to the red trajectories.

Using this result, we plot trajectories of $(A_1(t), A_2(t))$ in Fig. 1 for three values of ϵ. It is seen that for $\epsilon = 2.520 \times 10^{-3}$, the trajectory approaches the origin with no particularly notable features. By contrast, for $\epsilon = 2.294 \times 10^{-3}$, there appear two fixed points, and the trajectory approaches one of these. However, in this case, it eventually begins to move away from this point and converges to the origin in the $t \to \infty$ limit. As shown in the inset of Fig. 1, the part of the trajectory near the fixed point corresponds to the plateau in the graph of A_1 as a function of time. Finally, for $\epsilon = 2.285 \times 10^{-4}$, we find that the trajectory converges to the non-zero fixed point as $t \to \infty$. The appearance of the connection between this new fixed point and the origin is called a *saddle connection bifurcation* in dynamical system theory. The inset of Fig. 1 shows that the saddle connetion bifurcation in the two-dimensional system studied here corresponds to the non-ergodic transition of the time correlation function.

4. Concluding remark

We have elucidated the nature of a non-ergodic transition using a method of dynamical system reduction to treat the time correlation function. The key idea employed in our analysis is to focus on the zero eigenvectors $\mathbf{\Phi}_{00}$ and $\mathbf{\Phi}_{01}$ at the temperature T_0. Using these eigenvectors, we can obtain a simplified description of the large-dimensional dynamical system (12) in the form of the two-dimensional system represented by (20) and (21) near the temperature T_0. We find that the non-ergodic transition in the original system is manifested as the saddle connection bifurcation in the two-dimensional system. Before ending this paper, we make three important remarks.

First, our theoretical result for the system with the potential (3) has not been compared with numerical experiments yet. The comparison will be a future work. Here,

we note that our method can be applied to Langevin equations with other interaction potentials. As an example, for a model recently studied in Ref. [10], we confirmed the non-ergodic transition by both our method and a direct numerical experiment, though there is no precise quantitative correspondence between them. We also note that we do not understand the dependency of the results on potential forms.

The second remark is that (1) takes the same form as the density evolution equation in the over-damped limit obtained with the dynamical density functional method, where the potential function in (1) is replaced with the direct correlation function [6]. It is thus expected that our method can also be applied to this type of density evolution, though the temperature dependence becomes complicated.

Finally, recall that the equation we analyzed, (12), was obtained by ignoring fluctuations of $\Psi(\boldsymbol{k}, \boldsymbol{k}'; t, s)$. Thus, it is important to extend our treatment to one that accounts for fluctuations, in which (12) is derived as the zero-th order description. This problem is related to that of the growth of the length scale that appears near the glass transition point, which has been studied recently [11, 12]. We are now in the process of developing such a theory.

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